

# Evidence for weak itinerant long-range magnetic correlations in $\text{UGe}_2$

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Positive muon spin relaxation measurements performed on the ferromagnet  $\text{UGe}_2$  reveal, in addition to the well known localized  $5f$ -electron density responsible for the bulk magnetic properties, the existence of itinerant quasi-static magnetic correlations. Their critical dynamics is well described by the conventional dipolar Heisenberg model. These correlations involve small magnetic moments.

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The discovery of superconductivity below 1 K within a limited pressure range in the ferromagnet  $\text{UGe}_2$  [1–4] provides an unanticipated example of coexistence of superconductivity and strong ferromagnetism. The electronic pairing mechanism needed for superconductivity is believed to be magnetic in origin. However, it is amazing that ferromagnetically ordered uranium magnetic moments with so large magnitude ( $\sim 1.4 \mu_B$  at ambient pressure as deduced from magnetization measurements) are directly involved [5]. Since the pairing must involve the conduction electrons, it is important to characterize their magnetic properties. Because of the restrictions imposed by the magnetic form factor, this can not be done by diffraction techniques. As the muons localize in interstitial sites, they have the potentiality to yield information on the conduction electrons. Here we show, using the muon spin relaxation technique, that  $\text{UGe}_2$  is actually a dual system where two sub-states of  $f$  electrons coexist. We indeed report the existence at ambient pressure of itinerant long-range magnetic correlations with magnetic moments of  $\sim 0.02 \mu_B$  and a spectral weight in the megahertz range. A quantitative understanding of this state is moreover reached assuming that these correlations involve only long wavelength fluctuation modes.

$\text{UGe}_2$  is a ferromagnet with a Curie temperature  $T_C \simeq 52$  K which crystallizes in the orthorhombic  $\text{ZrGa}_2$  crystal structure (space group Cmmm) [6,7]. Magnetic measurements indicate a strong magnetocrystalline anisotropy [8,9,3] with easy magnetization axis along the  $\mathbf{a}$  axis.

We present results obtained by the muon spin relaxation ( $\mu\text{SR}$ ) technique. Fully polarized muons are implanted into the studied sample. Their spin ( $1/2$ ) evolves in the local magnetic field,  $\mathbf{B}_{\text{loc}}$ , until they decay into positrons. Since the positron is emitted preferentially in the direction of the muon spin at the decay time, it is possible to follow the evolution of the muon spin polarization [10,11]. The measured physical parameter is the so-called asymmetry which characterizes the anisotropy of the positron emission. Below  $T_C$ , if  $\mathbf{B}_{\text{loc}}$  has a compo-

nent perpendicular to the initial muon beam polarization,  $\mathbf{S}_\mu$  (taken parallel to  $Z$ ), we expect the asymmetry to display spontaneous oscillations with an amplitude maximum for  $\mathbf{B}_{\text{loc}} \perp \mathbf{S}_\mu$ . On the other hand, if  $\mathbf{B}_{\text{loc}} \parallel \mathbf{S}_\mu$ , the asymmetry can be written as the product of an initial asymmetry related to sample,  $a_s$ , and the muon spin relaxation function,  $P_Z(t)$ , which monitors the dynamics of  $\mathbf{B}_{\text{loc}}$ .

$\text{UGe}_2$  crystals were grown from a polycrystalline ingot using a Czochralski tri-arc technique [8]. We present results for two samples. Each consists of pieces cut from the crystals, put together to form a disk and glued on a silver backing plate. They differ by the orientation (either parallel or perpendicular) of the  $\mathbf{a}$  axis relative to the normal to the sample plane. The measurements were performed at the EMU spectrometer of the ISIS facility, from 5 K up to 200 K, mostly in zero-field. Additional  $\mu\text{SR}$  spectra were recorded with a longitudinal field.

We found that the temperature dependence of  $a_s$  for  $\mathbf{S}_\mu \parallel \mathbf{a}$  is consistent with  $\mathbf{B}_{\text{loc}} \parallel \mathbf{a}$ . In agreement with that conclusion, a spontaneous muon spin precession resulting in wiggles in the asymmetry is observed for  $\mathbf{S}_\mu \perp \mathbf{a}$ . Defining  $T_C$  as the temperature at which the wiggles disappear, we found  $T_C = 52.49$  (2) K. This value coincides with the maximum of the relaxation rate (to be evidenced below) for  $\mathbf{S}_\mu \parallel \mathbf{a}$  and  $\mathbf{S}_\mu \perp \mathbf{a}$ .

In this letter we focus on the description of data taken around the Curie point.

All the spectra were analyzed as a sum of two components:  $aP_Z^{\text{exp}}(t) = a_s P_Z(t) + a_{\text{bg}}$ . The first component describes the  $\mu\text{SR}$  signal from the sample and the second accounts for the muons stopped in the background, *i.e.* the cryostat walls and sample holder. In zero-field, for all relevant temperatures and for the two orientations of  $\mathbf{S}_\mu$  relative to  $\mathbf{a}$ ,  $P_Z(t)$  is well described by an exponential function:  $P_Z(t) = \exp(-\lambda_Z t)$  where  $\lambda_Z$  measures the spin-lattice relaxation rate at the muon site. An example is shown in Fig. 1.  $a_{\text{bg}}$ , which is basically temperature independent, was measured for  $\mathbf{S}_\mu \perp \mathbf{a}$  and  $T < T_C$  as

the constant background signal. We got  $a_{bg} = 0.077$  [12]. For  $\mathbf{S}_\mu \parallel \mathbf{a}$ , it could only be estimated from the sample size since the relaxation was never strong enough to measure it directly. We took  $a_{bg} = 0.064$ . The uncertainty on this  $a_{bg}$  leads to an uncertainty on the absolute value of  $\lambda_Z(\mathbf{S}_\mu \parallel \mathbf{a})$  of  $\sim 10\%$ .

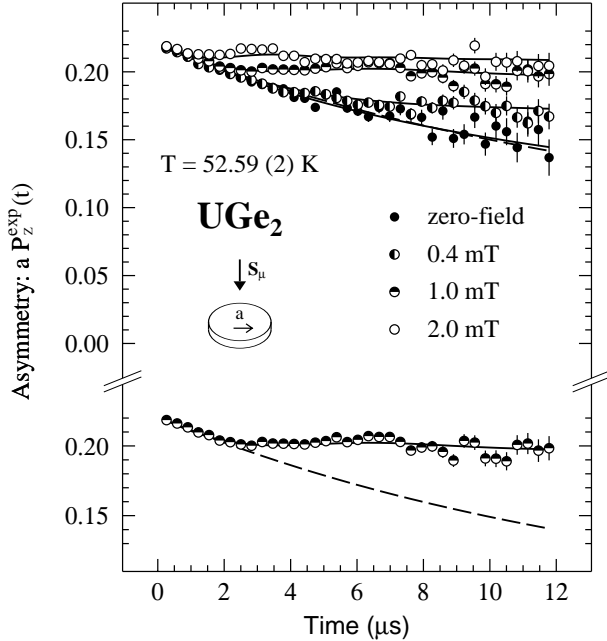


FIG. 1. Upper panel: examples of  $\mu$ SR spectra recorded in zero and longitudinal fields at  $T = 52.59$  (2) K (above  $T_C = 52.49$  (2) K) for  $\mathbf{S}_\mu \perp \mathbf{a}$ . The solid lines are fits assuming a squared-Lorentzian distribution for the modulus of the field at the muon site,  $B_{loc}$ . The dashed line, which is the result of the fit of the zero-field spectrum with an exponential relaxation function, can not be distinguished from the solid line except above  $\sim 11 \mu s$ . In the lower panel, the comparison of the 1.0 mT spectrum with the prediction of an exponential fit shows that this model is not valid in longitudinal fields. The field dependence at small field of  $P_Z(t)$  proves that the field distribution at the muon site is quasi-static.

In Fig. 2 we display  $\lambda_Z(T)$  measured in zero-field for  $\mathbf{S}_\mu \perp \mathbf{a}$  and  $\mathbf{S}_\mu \parallel \mathbf{a}$ . For both geometries,  $\lambda_Z(T)$  exhibits a maximum at  $T_C$ . It is due to the critical slowing down of the spin dynamics. Surprisingly the anisotropy between the orientations is very weak although  $UGe_2$  is known to be extremely anisotropic [14]. Furthermore we show in the following lines that  $\lambda_Z(T)$  near  $T_C$  is quantitatively understood in the framework of the Heisenberg model with dipolar interactions, whereas  $UGe_2$  is considered as an Ising system. The magnetic signal that we observe has therefore a different origin than the well documented uranium magnetic state observed e.g. by macroscopic measurements.

$\lambda_Z(T)$  has been computed several years ago [15] for the critical regime of dipolar Heisenberg ferromagnets and

has been successfully compared to experiments [15–17]. It is based on the derivation of the static and dynamical scaling laws from mode coupling theory [18]. The two scaling variables at play depend on two material parameters:  $\xi_0$ , the magnetic correlation length at  $T = 2T_C$ , and  $q_D$ , the dipolar wave vector which is a measure of the strength of the exchange interaction relative to the dipolar energy. This model initially derived for the paramagnetic phase applies also below  $T_C$  [16].

Specifically, the model predicts that  $\lambda_Z(T) = \mathcal{W}[a_L I^L(T) + a_T I^T(T)]$  where  $I^{L,T}$  [19] are scaling functions obtained from mode coupling theory and  $a_{L,T}$  are parameters determining respectively the amount of longitudinal (L) and transverse (T) fluctuations probed by the measurements. The L,T indices denote the orientation relative to the wave vector of the fluctuation mode.  $a_{L,T}$  only depend on muon site properties. The result of the fit of  $\lambda_Z(T)$  is shown in the inserts of Fig. 2. The divergence of  $\lambda_Z$  at  $T_C$  is strongly reduced by the effect of the dipolar interaction [18]. The temperature scale gives the product  $q_D \xi_0$  [15]. For  $\mathbf{S}_\mu \perp \mathbf{a}$ , we get  $q_D \xi_0 = 0.021$  (2), and for  $\mathbf{S}_\mu \parallel \mathbf{a}$ ,  $q_D \xi_0^+ = 0.043$  (2) and  $q_D \xi_0^- = 0.020$  (2). The index + (−) on  $\xi_0$  specifies that we consider the paramagnetic (ferromagnetic) state.  $\xi_0(\mathbf{S}_\mu \parallel \mathbf{a}) > \xi_0(\mathbf{S}_\mu \perp \mathbf{a})$  in the paramagnetic state, suggesting that the magnetic correlations are somewhat anisotropic. The fact that  $\xi_0^+ > \xi_0^-$  is an expected feature [10]. The relaxation rate scale yields  $\mathcal{W}^+ a_L = 0.140$  (4) MHz and  $\mathcal{W}^- a_L = 0.20$  (2) MHz for  $\mathbf{S}_\mu \parallel \mathbf{a}$ . The transverse contribution to  $\lambda_Z$  for both  $T < T_C$  and  $T > T_C$  is more difficult to estimate since  $a_T$  is found much lower than  $a_L$ . Reasonable fits are obtained with  $a_T/a_L = 0.036$  (14). We have computed  $a_L$  and  $a_T$  for different possible muon sites and found only one site satisfying  $a_T < a_L/2$ . This is site 2b (in Wyckoff notation) of coordinates (0, 1/2, 0) for which  $a_L = 1.2486$ ,  $a_T = 0.0386$ . We then deduce  $\mathcal{W}^+ = 0.112$  (3) MHz and  $\mathcal{W}^- = 0.161$  (16) MHz. The scale deduced from the measurements with  $\mathbf{S}_\mu \perp \mathbf{a}$  is about twice as large, pointing out again to the weak anisotropy of the magnetic correlations.

In order to further characterize the relaxation near  $T_C$ , we performed at a given temperature longitudinal field measurements for the two orientations of  $\mathbf{S}_\mu$  relative to  $\mathbf{a}$ . The field responses for the two geometries are similar. An illustration is given in Fig. 1. Surprisingly, the spectra are field dependent at extremely low  $B_{ext}$ , proving that the probed magnetic fluctuations are quasi-static (fluctuation rate in the MHz range) and since  $\lambda_Z$  is small, the associated magnetic moment must be small as well. Quantitatively, the field dependence of  $P_Z(t)$  can not be described consistently either by a simple exponential relaxation form (see the lower panel of Fig. 1) nor by a relaxation function computed with the strong collision model assuming an isotropic Gaussian component field distribution [20]. On the other hand, the relaxation is well explained if we assume that the distribution of the

local field at the muon,  $B_{\text{loc}}$ , is squared-Lorentzian [21]. We write  $P_Z(t) = P_Z(\Delta_{\text{Lor}}, \nu_f, t)$  where  $\Delta_{\text{Lor}}$  characterizes the width of the field distribution and  $\nu_f$  its fluctuation rate [22]. A global fit of the spectra ( $B_{\text{ext}} = 0, 0.2, 0.4, 0.6, 0.8, 1.0$  and  $2.0$  mT) taken at a given temperature is possible. For  $\mathbf{S}_\mu \perp \mathbf{a}$  at  $T = 52.59$  (2) K, the description of the seven spectra is done with  $\Delta_{\text{Lor}} = 70$   $\mu\text{T}$  and  $\nu_f = 0.10$  MHz. For  $\mathbf{S}_\mu \parallel \mathbf{a}$  at  $T = 52.47$  (2) K the two parameters are  $\Delta_{\text{Lor}} = 40$   $\mu\text{T}$  and  $\nu_f = 0.50$  MHz: the zero-field spectra have therefore been recorded in the motional narrowing limit ( $\nu_f/(\gamma_\mu \Delta_{\text{Lor}}) > 1$  where  $\gamma_\mu$  is the muon gyromagnetic ratio;  $\gamma_\mu = 851.6$  Mrad s $^{-1}$ T $^{-1}$ ). This justifies the formalism used to treat  $\lambda_Z(T)$  close to  $T_C$ .

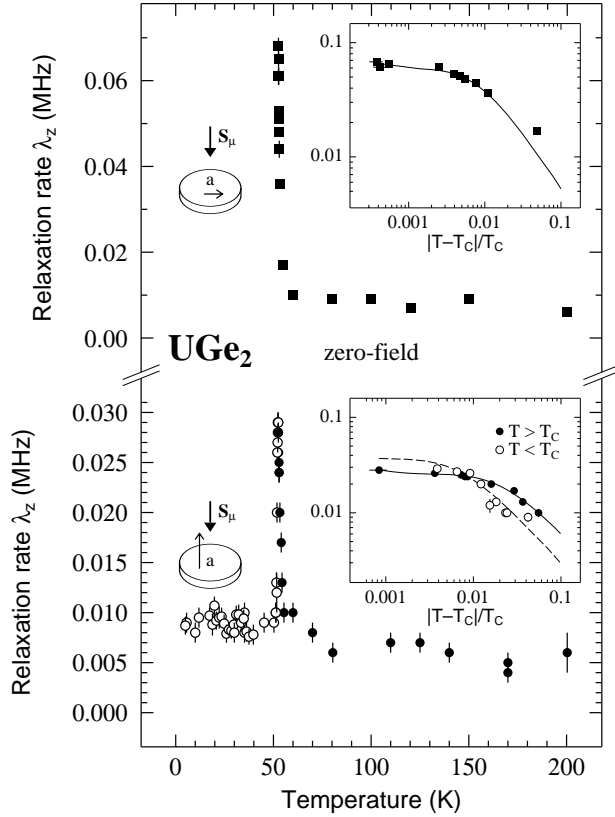


FIG. 2. Temperature dependence of  $\lambda_Z$  measured in zero-field for  $\mathbf{S}_\mu \perp \mathbf{a}$  and  $\mathbf{S}_\mu \parallel \mathbf{a}$  in the upper and lower panels, respectively. The inserts display  $\lambda_Z(T)$  near  $T_C$ . The solid and dashed lines are the results of fits for a dipolar Heisenberg ferromagnet as explained in the main text. Since  $\mathbf{B}_{\text{loc}} \parallel \mathbf{a}$ , we can not observe a spin-lattice relaxation process for  $\mathbf{S}_\mu \perp \mathbf{a}$  in the ordered state. The point for  $\mathbf{S}_\mu \perp \mathbf{a}$  at  $(T - T_C)/T_C = 0.05$  does not fit the critical description, pointing out that it was recorded outside the critical region for that geometrical configuration. The behavior of  $\lambda_Z(T)$  near  $T_C$  is typical for a ferromagnet in its critical regime.

We now present an interpretation of our results.

We first note that the detected fluctuations can not

arise directly from the localized uranium  $5f$  electrons since  $\nu_f$  would then be in the THz window as estimated from  $\nu_f \simeq k_B T_C / \hbar$ , rather than in the MHz range as measured. We also already mentioned that the observed  $\mu\text{SR}$  signal has not the properties expected from the known macroscopic properties. These apparently conflicting results can be understood if the  $5f$  electrons are viewed as two electron subsets. This picture has already been argued for  $\text{UCu}_5$  [23] and  $\text{UPd}_2\text{Al}_3$  [24–28]. However, for  $\text{UGe}_2$  the signatures of both subsets are found at a single temperature, the Curie temperature, whereas for  $\text{UCu}_5$  and  $\text{UPd}_2\text{Al}_3$  the temperatures at which the two subsets are detected are far apart. So  $\text{UGe}_2$  presents a novel variant of the two electron subset model. Within this picture, the anisotropy of the magnetization arises from the localized  $5f$  spectral density and the magnetic fluctuations probed by  $\mu\text{SR}$  is a signature of the band-like electrons. We do not detect the signature of the localized  $5f$  electrons, because of the strong motional narrowing of the related relaxation rate. It is observed for the ferromagnet  $\text{YbNiSn}$ ; see Ref. [13].

The effect of the dipolar interaction on the quasi-elastic linewidth,  $\Gamma(q)$ , of the fluctuations has already been observed for the weak itinerant ferromagnet  $\text{Ni}_3\text{Al}$  [29]. In particular, at criticality  $\Gamma(q) \propto q^{5/2}$ , as expected from scaling [18]. Thus it is not completely surprising to detect its influence on  $\lambda_Z(T)$  for the band-like electrons of  $\text{UGe}_2$ . Quantitatively, the data have been described in the established framework of critical dynamics [18]. We shall now prove that the detected magnetization density arises entirely from long wavelength, i.e. small  $q$ , fluctuations. The magnetic properties of weak itinerant ferromagnets are explained with the latter hypothesis [30,31]. In our model the values of  $\mathcal{W}$  and  $\nu_f$  and of the magnitude of the band-like uranium magnetic moment,  $m_U$ , are controlled by two wavevectors:  $q_D$  already introduced and the cut-off wavevector,  $q_c$ , which sets the upper bound for the wavevector of the fluctuations involved in the build-up of the magnetization density. For simplicity we consider that the magnetic properties of this electronic subset are isotropic. We shall detail the analysis of the data taken with  $\mathbf{S}_\mu \parallel \mathbf{a}$ . The same approach works equally well for the data recorded with  $\mathbf{S}_\mu \perp \mathbf{a}$ . As explained below, we get an overall consistent picture setting  $q_D = 1.0 \times 10^{-3}$   $\text{\AA}^{-1}$  and  $q_c = 0.1$   $\text{\AA}^{-1}$ .

The magnetization arising from the conduction electrons can be viewed as a stochastic variable with a variance  $\langle (\delta \mathcal{M})^2 \rangle$ . From the fluctuation-dissipation (Nyquist's) theorem [32],  $\langle (\delta \mathcal{M})^2 \rangle$  obeys the sum rule

$$\langle (\delta \mathcal{M})^2 \rangle = \frac{3 k_B T}{2 \pi^2 \mu_0} \int_0^{q_c} \chi(q) q^2 dq, \quad (1)$$

if the energy of the magnetic fluctuations is smaller than the thermal energy.  $\mu_0$  is the permeability of free space. Assuming an Ornstein-Zernike form for the wavevector dependent susceptibility,  $\chi(q)$ , and since  $q_D$

is very small,  $\langle(\delta\mathcal{M})^2\rangle \simeq 3k_{\text{B}}Tq_{\text{D}}^2q_{\text{c}}/(2\pi^2\mu_0)$ . Since  $m_{\text{U}} = v_0\sqrt{\langle(\delta\mathcal{M})^2\rangle}$  where  $v_0$  is the volume per uranium atom ( $v_0 = 61.6 \text{ \AA}^3$ ), we infer  $m_{\text{U}} = 0.02 \mu_{\text{B}}$  at  $T_{\text{C}}$ . Interestingly, the analysis of polarized neutron scattering data suggests for the conduction electrons a magnetic moment of  $0.04(3) \mu_{\text{B}}$  at low temperature [33].

The scale  $\mathcal{W}$  for  $\lambda_{\text{Z}}$  can then be computed within the framework presented above. Numerically, from Eq. 5.10c of Ref. [15], we get  $\mathcal{W} = 0.16 \text{ MHz}$ , close to the measured values. With the same theory,  $\hbar\Gamma(q) = \Omega q^{5/2}$  with  $\Omega = 18 \text{ meV \AA}^{2.5}$  at criticality and for small  $q_{\text{D}}$  (see Eq. 4.14b of Ref. [15]). Since the measured dynamics is mainly driven by the fluctuations at  $q_{\text{D}}$  [19], we estimate  $\nu_f \simeq \Gamma(q_{\text{D}}) = 0.87 \text{ MHz}$ , not far from the measured value.

We now discuss the magnitude of  $\Delta_{\text{Lor}}$ . If the distribution of  $B_{\text{loc}}$  was Gaussian, the zero-field width of the distribution would be  $\Delta_{\text{Gauss}} = 1.7 \text{ mT}$  for muon at site  $2b$ , and  $m_{\text{U}} = 0.02 \mu_{\text{B}}$  computed using the Van Vleck type formalism of Ref. [20]. However the distribution is squared-Lorentzian rather than Gaussian. Such a distribution is observed in systems with diluted and disordered magnetic moments [21]. According to Uemura *et al.* [22],  $\Delta_{\text{Lor}} = \sqrt{\pi/2} c \Delta_{\text{Gauss}}$  where  $c$  is the concentration of moments at the origin of the distribution. This relation leads to  $c = 1.9 \%$ , consistent with the usual fact that a tiny fraction of the total number of valence electrons are able to contribute to the magnetic susceptibility.

From the  $q_{\text{D}}$  value we derive the exchange interaction. We obtain  $2\mathcal{J} = k_{\text{B}}T_{\text{C}}/4.2$  (see Eq. 4.4b of Ref. [15] and Ref. [18]). For comparison, the same method gives  $2\mathcal{J}/k_{\text{B}}T_{\text{C}} = 1/11$  and  $1/20$  for metallic Fe and Ni, respectively. Therefore the evaluation of the exchange energy is quite reasonable. From the measured product  $q_{\text{D}}\xi_0^+$ , we get  $\xi_0^+ \simeq 43 \text{ \AA}$ . This means that the interaction between the itinerant magnetic moments is relatively long-range, even far outside the critical regime. Although about an order of magnitude larger than for conventional ferromagnets,  $\xi_0^+$  compares favorably with the neutron result for  $\text{Ni}_3\text{Al}$ :  $\xi_0^+ = 24(9) \text{ \AA}$  [29]. For the same compound, we derive from [30] that  $q_{\text{c}} = 0.2 \text{ \AA}^{-1}$ , a value twice as large as found for  $\text{UGe}_2$ . The moment carried by the itinerant electrons is about four times smaller for  $\text{UGe}_2$  than for  $\text{Ni}_3\text{Al}$  [34]. Nearest neighbor U atoms form zigzag chains parallel to  $\mathbf{a}$  [3]. This may lead to magnetic frustration and thus explains the disordered nature of the distribution of  $B_{\text{loc}}$ . A proper understanding of the origin of the squared-Lorentzian distribution requires more work.

One may question the uniqueness of our interpretation. We first note that the observed  $\lambda_{\text{Z}}$  can not arise from an impurity phase since the measured critical dynamics occurs right at the well known Curie temperature of  $\text{UGe}_2$ . It could be argued that the observed signal is the signature of a weak disorder in the uranium magnetic moments. This has already been seen in UAs [35] where

the  $\mu\text{SR}$  signal below the Néel temperature has been attributed to a diluted source of small magnetic moments. Their quasi-static nature is related to the absence of spin excitations. However, the moments we observe in  $\text{UGe}_2$  are quasi-static even above  $T_{\text{C}}$ .

In conclusion we have shown that at ambient pressure  $\text{UGe}_2$  is a dual system where an electronic subset of itinerant states coexist with the subset of localized  $5f$  electrons responsible for the well known bulk magnetic properties. Its associated magnetic moment is quite small and characterized by a very slow spin dynamics. A quantitative picture for that subset is achieved by assuming that only fluctuations at long wavelength are at play. It would be of interest to follow the small moment itinerant state as a function of pressure to determine whether the Cooper's pairs arise from it. However, it seems difficult to perform that task with  $\mu\text{SR}$ , unless the spin-lattice relaxation rate increases appreciably at high pressure.

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